

Argonne National Laboratory

**FISSION PRODUCT MONITORING IN
EBR-I, MARK IV**

by

R. R. Smith and C. B. Doe

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I. INTRODUCTION

In the event of a failure of a fuel element cladding in EBR-I, Mark IV, minute quantities of fission products will be released directly to the NaK coolant. For a minor failure, there is no compelling reason why normal operations should be interrupted. Since the primary NaK coolant is normally highly radioactive (from Na^{24}), the contribution to the specific activity of the coolant from a credible fission product release will be essentially immeasurable. All primary components are heavily shielded and are inaccessible during and following (for approximately one week) a normal operating period. In principle, the reactor could be operated indefinitely with essentially no aggravation of personnel-monitoring problems.

For a serious failure or for a series of small failures, however, continued operation may lead to a fission product accumulation sufficiently large to affect the accessibility of primary components for occasional maintenance.⁽¹⁾ From the operational point of view, some system capable of annunciating and analyzing the seriousness of a cladding failure is a necessity. On the basis of information gleaned from such a system, the operator may either elect to continue operation or to shut down, locate, and replace the damaged rod. This report describes the development and testing of a monitoring system capable of yielding such information.

II. DESCRIPTION OF THE MONITORING PRINCIPLE

For reactor systems in which the coolant is water or a gas such as helium, carbon dioxide, or air, the release of fission products from the fuel may be annunciated by an activity monitor installed at some downstream location. Interference from activities generated in such coolants is usually either of minor significance or can be satisfactorily reduced by relatively simple methods of discrimination.⁽²⁾

If the coolant is sodium or a sodium-potassium alloy (such as NaK), the problem of annunciation is more complicated, since the monitor must be capable of detecting a small fission product signal in the presence of an overriding background originating from the decay of Na^{24} which is generated

continuously in the coolant. The quality of information afforded by any conceivable device designed to separate a true fission product signal from an overriding Na^{24} activity is certain to be questionable.

The necessity of blanketing the primary coolant in a fast reactor (usually sodium or NaK) with an inert gas such as argon, however, suggests an interesting possibility, since it is known that rare gas fission products (krypton and xenon) diffuse rapidly into the cover gas system.⁽³⁾ In principle, some suitable beta-gamma detector which monitors the activity of the cover gas should be capable of announcing a cladding failure. In practice, it is necessary to effect an almost complete discrimination against A^{41} , which is present in the cover gas at activity levels many orders of magnitude higher than those associated with credible concentrations of rare gas fission products.

A simple and extremely efficient method of discrimination against A^{41} has been developed, a method which utilizes the selective electrostatic fixation of rare gas daughter nuclides.⁽⁴⁾ The principle of the fixation process is premised on the fact that at the instant of beta decay, each of the daughter species becomes a positively charged ion. Accordingly, the cover gas is passed through an electrostatic precipitation unit which consists primarily of a cylinder maintained at a potential of 2500 V positive. A closed-loop wire driven axially through the cylinder is maintained at ground. Upon beta decay, the ionized daughter species migrate to the wire and become electronically neutralized. The wire carrying the neutralized daughters passes through a mercury trap-type gas seal which has a few milliliters of water floating on its surface. The strongly electropositive rubidium and cesium daughters react instantly with the water and become permanently fixed in the water layer in the hydroxide form. The isotope K^{41} , the daughter of A^{41} , also reacts with the water, but since K^{41} is radioactively inert, the activity associated with the water fraction is almost exclusively that arising from the decay of various rubidium and cesium fission products. It follows that the output from a detector which monitors the activity level of the water fraction may be used as a criterion of the fission product concentration in the cover gas.

III. EQUIPMENT

The principal item of interest is the electrostatic precipitation chamber, illustrated schematically in Fig. 1. The unit consists essentially of two concentric cylinders: an inner, 2 in. in diameter, of aluminum; and an outer, 4 in. in diameter, of stainless steel. Running axially through the inner cylinder (the anode) is a 6-mil, closed-loop, stainless steel wire driven at a speed of approximately 18 in./min by a 3-rpm Bodine induction motor. High voltage (2500 V positive) is supplied to the anode through a

conventional Teflon-insulated high-voltage fitting. Plexiglas end caps (not illustrated) insulate the anode from the outer cylinder. The remainder of the precipitation unit, including the collection wire, is maintained at ground potential.

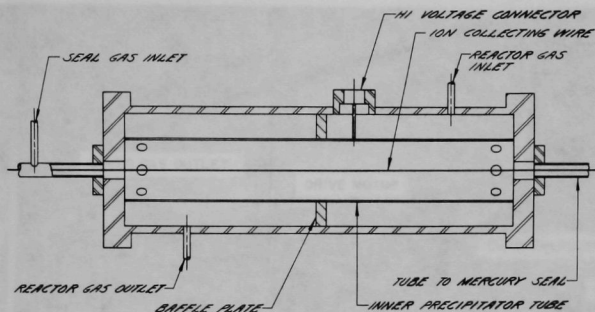


Fig. 1. Precipitation Chamber for Cladding Failure Monitor

Cover gas at less than 5 psi and flowing at a rate of approximately 100 ml/min is admitted to the chamber through $\frac{1}{4}$ -in. copper tubing and is exhausted at near-atmospheric pressure to the reactor stack system. Cover gas enters and leaves the cylindrical anode through $\frac{1}{4}$ -in. holes drilled at opposite ends. Short-circuit gas flow between inlet and outlet ports is prevented by a Plexiglas baffle plate.

To avoid the leakage of active cover gas to the surroundings from the wire inlet and outlet ports, two mercury seals in the form of U-tubes are used. One of these, along with other components, is shown in Fig. 2; the other, the downstream trap, is located behind lead shielding bricks. Floating on the surface of the mercury in the upstream leg of the downstream trap is a layer of water approximately 1 in. in depth. The water, through its chemical reaction with the rubidium and cesium daughters, serves two purposes: to integrate the activity with respect to time, and to decontaminate the wire continuously. A system of pulleys in each U-tube enables the moving wire to negotiate the necessary bends. Two idler pulleys supply tension to the wire.

To prevent active cover gas from diffusing into the downstream trap, the region between the water layer and the precipitation unit is flushed continuously under positive pressure with uncontaminated argon from the reactor cover gas feed system. Flowmeters and valves installed in the flush and active cover gas inlet lines serve as a means of flow control.

The detector used to monitor the various activities collected in the water fraction is a 2 x 2 in. NaI (Tl-activated) crystal optically coupled to

a Dumont 8219 photomultiplier tube. To reduce the effects of radiations emitted from the reactor and from the precipitation chamber, the crystal and photomultiplier tube assembly are enclosed in a cylindrical lead shield approximately 4 in. in thickness.

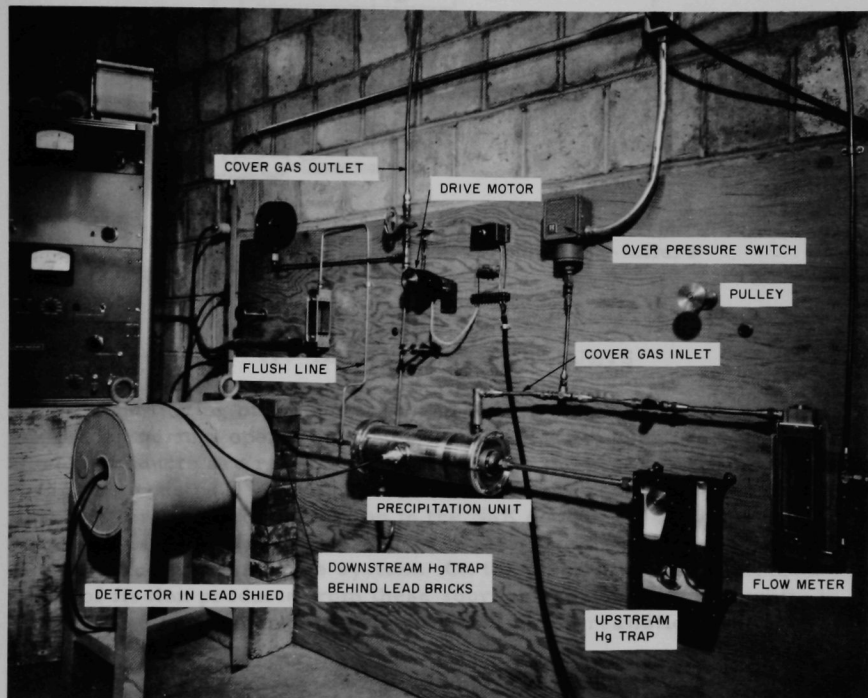


Fig. 2. Photograph of the Working Monitor Unit

During earlier stages of the tests, the output from the photomultiplier tube was fed to a Nuclear Data 256-channel transistorized pulse-height analyzer equipped with both photo-oscillographic and digital readouts. Periodic measurements with Cs^{137} and Co^{60} sources were used to establish the energy calibration of the spectrometer.

In later stages of the tests, after the feasibility of the method was established, the spectrometer was replaced with more conventional equipment. Pulses from the photomultiplier tube were amplified by a linear amplifier and fed to a conventional count rate meter. A block diagram of the various components is given in Fig. 3.

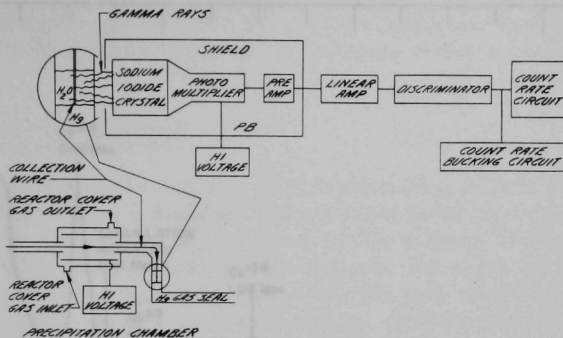


Fig. 3. Block Diagram for Cladding-failure Monitor

IV. EXPERIMENTAL RESULTS

The testing of the equipment in EBR-I was complicated by the fact that under normal operating conditions, substantial quantities of rare gas fission products are released continuously to the cover gas from the following sources: (1) vented blanket thermocouple rods, and (2) a small, unknown, uranium contamination in the NaK coolant.⁽¹⁾ Obviously, the natural existence of fission products in the system reduces the sensitivity and the effectiveness of the system. On the other hand, the continuous release of fission products from these sources provided an opportunity to evaluate the performance of the equipment under realistic conditions of fission product release.

A. Discrimination against A^{41}

In a system such as the one described above, the possibility exists that A^{41} may be carried over into the downstream (monitored) trap, either through diffusion, sorption into the stainless steel wire, or through some unsuspected mechanism. To test for this possibility and to assess the degree of discrimination against A^{41} , 0.1 ml of water was taken from the trap after 2 hr of full-power operation and analyzed with a Nuclear Data 256-channel pulse-height analyzer. The results are given in Fig. 4. Each of the gamma peaks may be identified with radiations associated with specific isotopes of rubidium and cesium. From the spectrum, it is clear that the 1.29 MeV gamma of A^{41} is almost completely missing. In contrast, a pulse-height analysis of an active cover gas sample (not illustrated) reveals the existence of a single peak at 1.29 MeV. Radiations from rare gas fission products and their respective daughters, while present, are completely masked by the much higher activity associated with A^{41} . It is clear from these considerations that the degree of discrimination against A^{41} is satisfactorily high.

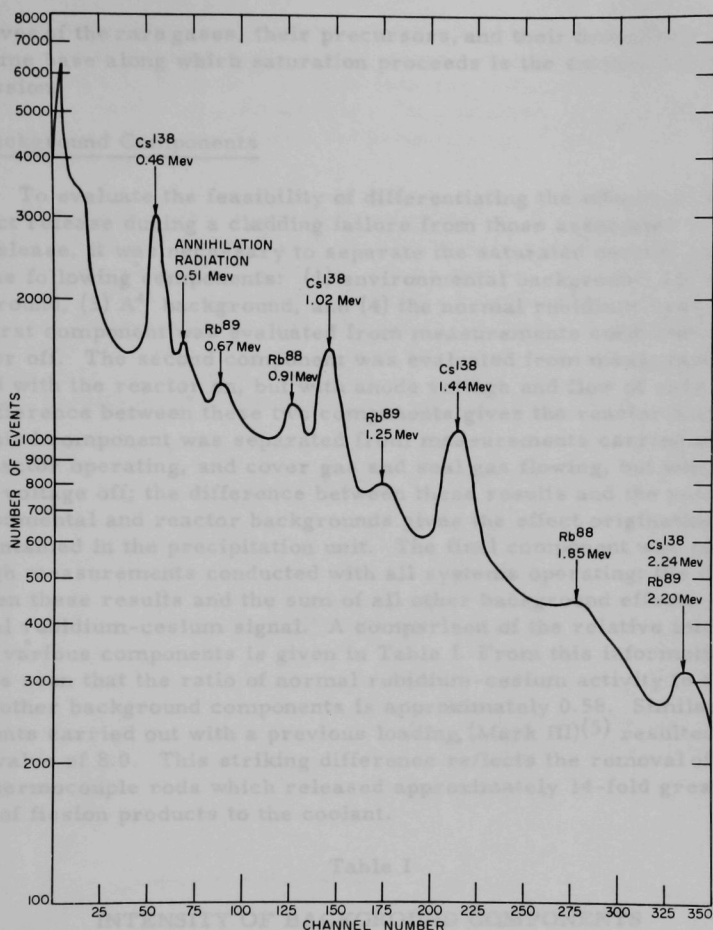


Fig. 4. Gamma Pulse-Height Spectrum of Water Fraction

As the spectrum of Fig. 4 indicates, essentially all of the activity collected in the water fraction is the result of decay of three fission product species, i.e., Rb⁸⁸, Rb⁸⁹, and Cs¹³⁸. The half-lives of each of these species are short, being respectively 18, 15, and 32 min. In addition, the respective half-lives of their precursors are also short, i.e., 2.8 hr, 3.2 min, and 32 min. It follows that the decay of precursors and daughters will be essentially complete for shutdown periods longer than a few hours. During a startup following a prolonged shutdown (overnight, for example), it is clear that the activity level of the water fraction will increase rapidly from zero and will eventually reach equilibrium at a time dictated by the

half-lives of the rare gases, their precursors, and their immediate daughters. The time base along which saturation proceeds is the subject of a later discussion.

B. Background Components

To evaluate the feasibility of differentiating the effects of fission product release during a cladding failure from those associated with normal release, it was necessary to separate the saturated normal response into the following components: (1) environmental background, (2) reactor background, (3) A^{41} background, and (4) the normal rubidium-cesium signal. The first component was evaluated from measurements conducted with the reactor off. The second component was evaluated from measurements conducted with the reactor on, but with anode voltage and flow of cover gas off; the difference between these two components gives the reactor background. The third component was separated from measurements carried out with the reactor operating, and cover gas and seal gas flowing, but with the anode voltage off; the difference between these results and the sum of the environmental and reactor backgrounds gives the effect originating from A^{41} contained in the precipitation unit. The final component was established through measurements conducted with all systems operating; the difference between these results and the sum of all other background effects gives the normal rubidium-cesium signal. A comparison of the relative intensities of the various components is given in Table I. From this information, it may be seen that the ratio of normal rubidium-cesium activity to the sum of all other background components is approximately 0.58. Similar measurements carried out with a previous loading (Mark III)⁽⁵⁾ resulted in a ratio value of 8.0. This striking difference reflects the removal of 12 vented fuel thermocouple rods which released approximately 14-fold greater quantities of fission products to the coolant.

Table I

INTENSITY OF BACKGROUND COMPONENTS IN EBR-I, MARK IV

Component	Source	Intensity (cps)
1	Environmental	4
2	Reactor	36
3	A^{41}	0
4	Rb-Cs	23
Total	All sources	63

C. Simulated Failure of Fuel Rod

To obtain some realistic measure of the time response and the strength of the signal resulting from a failure of fuel rod cladding, a

simulation experiment was carried out with the Mark IV loading. Twenty unclad enriched uranium foils, each 1 x 1 cm, were packed into a well-ventilated (with NaK) basket which in turn was installed near the edge of the core in a NaK-ventilated thimble. The combined area of the twenty foils was chosen to coincide with the exposed area of fuel in a typical Mark IV fuel rod.

The results of measurements conducted at 900 kW (full power, 1200 kW) are illustrated in Fig. 5, which gives the activity associated with the water layer as a function of time after reaching operating power. The lower curve, obtained under normal operating conditions, gives a true measure of the background level. As expected, the activity, extremely low shortly after startup, increases rapidly and eventually reaches saturation approximately 3 hr after startup. Repeat measurements conducted on three successive days demonstrated the reproducibility of the results.

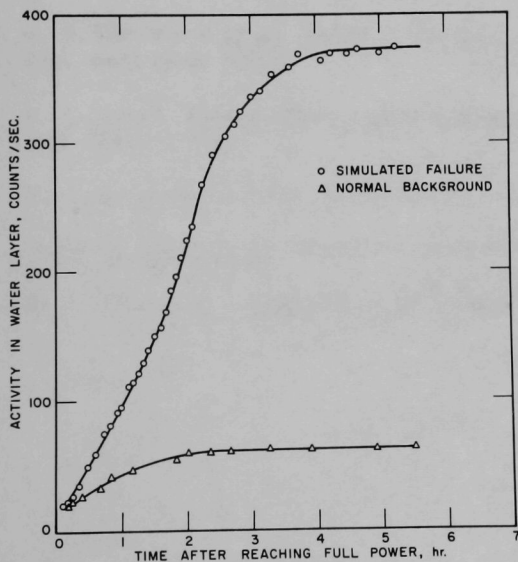


Fig. 5

Fuel Rod Failure Simulation, EBR-I, Mark IV

The upper curve of Fig. 5 gives the results of measurements carried out with the unclad enriched uranium foils in position and with all other conditions unchanged. Hence, for any given time the intensity difference between the two curves gives a measure of the fission product release from the unclad foils. To permit a reasonable evaluation of the effect expected from the failure of a single Mark-IV fuel rod, corrections for the difference between Pu^{239} and U^{235} fission cross sections have been applied to the data illustrated.

From the results of Fig. 5, it is clear that the strength of the fission product signal from the simulated failure is approximately five times that of the normal background signal. It is reasonable to conclude from the results of this somewhat idealized experiment that the release of fission products from failure of a Mark-IV fuel rod cladding would be detectable.

In conclusion, it seems clear that the charged-wire cover gas monitoring principle may find useful application in other sodium-cooled fast reactor systems.

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